1T-TaS_2 as a quantum spin liquid

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1T-TaS_2 is unique among transition metal dichalcogenides in that it is understood to be a correlation-driven insulator, where the unpaired electron in a 13-site cluster experiences enough correlation to form a Mott insulator. We argue, based on existing data, that this well-known material should be considered as a quantum spin liquid, either a fully gapped *Z2* spin liquid or a Dirac spin liquid. We discuss the exotic states that emerge upon doping and propose further experimental probes.

spin liquid | Mott insulator | transition metal dichalcogenide

The transition metal dichalcogenide (TMD) is an old subject that has enjoyed a revival recently due to the interests in its topological properties and unusual superconductivity. The layer structure is easy to cleave or intercalate and can exist in single-layer form (1, 2). This material was studied intensively in the 1970s and 1980s and was considered the prototypical example of a charge density wave (CDW) system (3). Due to imperfect nesting in two dimensions, in most of these materials, the onset of CDW gaps out only part of the Fermi surface, leaving behind a metallic state that often becomes superconducting. Conventional band theory and electron–phonon coupling appear to account for the qualitative behavior (3). There is, however, one notable exception, namely 1T-TaS_2. The Ta forms a triangular lattice, sandwiched between two triangular layers of S, forming an ABC-type stacking. As a result, the Ta is surrounded by S, forming an approximate octahedron. In contrast, the 2H-TaS_2 forms an ABA-type stacking, and the Ta is surrounded by S, forming a trigonal prism. In a single layer, inversion symmetry is broken in 2H structure. The system is a good metal below the CDW onset around 90 K, and, eventually, the spin–orbit coupling gives rise to a special kind of superconductivity called Ising superconductivity (4–6). In 1T-TaS_2, inversion symmetry is preserved. The system undergoes a CDW transition at about 350 K with a jump in the resistivity. It is known that this transition is driven by an incommensurate triple-Q CDW (ICDW). A similar transition is seen in 1T-$TaSe_2$ at 470 K. However, whereas $TaSe_2$ stays metallic below this transition, 1T-TaS_2 exhibits a further resistivity jump around 200 K that is hysteretic, indicative of the first-order nature of this transition. These transitions are also visible in the spin susceptibility data shown in Fig. 2. In early samples, the resistivity rises only by about a factor of 10 as the temperature is lowered from 200 K to 2 K and, below that, obeys Mott hopping law (log resistivity goes as $T^{-1/3}$) (7). More recent samples show better insulating behavior, and it is generally agreed that the ground state is insulating. The 200 K transition is accompanied by a lock-in to a commensurate CDW (CCDW), forming a $\sqrt{13} \times \sqrt{13}$ structure. As shown in Fig. 1, this structure is described as clusters of stars of David where the sites of the stars move inward toward the site in the middle. The stars of David are packed in such a way that they form a triangular lattice. Thus, the unit cell is enlarged to have 13 Ta sites. The formal valence of Ta is 4+, and each Ta site has a single $5d$ electron. We have an odd number of electrons per unit cell. (We first restrict ourselves to a single layer. Interlayer effects will be discussed later in this section.) According to band theory, the ground state must be metallic. The only option for an insulating ground state in the pure material is a correlation driven Mott insulator. This fact was pointed out by Fazekas and Tosatti (8) in 1976. Band calculations show that band folding creates a cluster of bands near the Fermi surface. Rossnagel and Smith (9) found that, due to spin–orbit interaction, a very narrow band is split off, which crosses the Fermi level with a 0.1- to 0.2-eV gap to the other subbands. The narrow bandwidth means that a weak residual repulsion is sufficient to form a Mott insulator, thus supporting the proposal of Fazekas and Tosatti, and distinguishes 1T-TaS_2 from other TMD. Apparently, the formation of the commensurate clusters is essential for the strong correlation behavior in these $4d$ and $5d$ systems. Currently, the assignment of cluster Mott insulator to the undoped 1T-TaS_2 ground state is widely accepted. A band about 0.2 eV below the Fermi energy has been interpreted as the lower Hubbard band in angle-resolved photo-emission spectroscopy (ARPES) (10, 11), and the electronic-driven nature of the 200 K transition has been confirmed by time-dependent ARPES on the basis of the fast relaxation time of the spectra (12).

Surprisingly, with a few exceptions, the issue of magnetism associated with the Mott insulator has not been discussed in the literature. In the standard picture of a Mott insulator, the spins form local moments, which then form an antiferromagnetic (AF) ground state due to exchange coupling. No such AF ordering has been reported in 1T-TaS_2. There is not even any sign of the local moment formation, which usually appears as a rise in the spin susceptibility with decreasing temperature, following a Curie Weiss law. As seen in Fig. 2, the magnetic susceptibility drops at the CDW transitions, but remains almost flat below 200 K (3). (The small rise below 50 K can be attributed to 5×10^{-4} impurity spin per Ta.) This serious discrepancy from the Mott picture did not escape the attention of Fazekas and Tosatti (8); they attempted to explain this by arguing that the g factor is very small due to spin–orbit coupling. However, their argument depends sensitively on the assumption of a cubic environment for the Ta atoms, which is now known not to be true. In fact, Rossnagel and Smith (9) claim that a single band crosses the Fermi level that is mainly made up of xy and $x^2 - y^2$ orbitals. This picture suggests

Significance

In solids with an odd number of electrons per unit cell, band theory requires that they are metals, but strong interaction can turn them into insulators, called Mott insulators. In this case, the electrons form local moments that, in turn, form an antiferromagnetic ground state. In 1973, P. W. Anderson proposed that, in certain cases, quantum fluctuations may prevent magnetic order and result in a paramagnetic ground state called a quantum spin liquid. After many years of searching, a few examples have been discovered in the past several years. We point out that a well-studied material, TaS_2, may be a spin liquid candidate. We propose further experiments that probe the exotic properties of this new state of matter.

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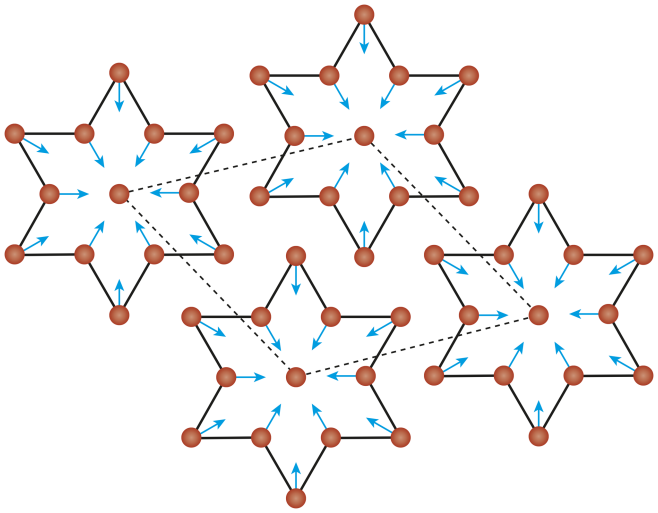

Fig. 1. In the cluster Mott phase of 1T-TaS_2, Ta atoms (red dots) belonging to a star of David move toward the Ta atom at the center. Thirteen Ta atoms form a unit cell, and these unit cells form a triangular lattice. The directions and lengths of the arrows are schematic.

that the low-energy physics can be described by a single-band Hubbard model. Due to the strong spin–orbit coupling, spin is not a good quantum number, but there is a pseudospin made up of Kramers pair to describe the degeneracy of each band in the presence of inversion symmetry. Throughout this paper, spin will refer to this pseudospin. In general, there is no $SU(2)$ symmetry for the pseudospin, and we expect a nonzero and anisotropic g factor. Thus, we believe we cannot rely on the Fazekas–Tosatti proposal to explain the absence of Curie Weiss behavior.

A more recent discussion on magnetism came in 2005, when Perfetti et al. (10) proposed the existence of a fluctuating spin density wave (SDW) to explain certain features in their ARPES data. In the CCDW phase, they reported a band about 0.15 eV below the Fermi energy, which has a small upward dispersion. In addition, they reported spectral weight above this band, consistent with a back-folded band with a downward dispersion, which is greatly smeared. They compared this observation with the dispersion calculated for a triple-Q SDW and argued that their observation supports some kind of incipient SDW order. The spin density of this SDW is smoothly connected to that of the three-sublattice 120° spin order of local moments expected for the Heisenberg model on a triangular lattice. Presumably, the SDW fails to order due to quantum fluctuation. If so, the picture presented in ref. 10 is smoothly connected to a spin liquid state formed out of local moments, even though there is no sign of local moments from the SDW picture. It should be pointed out, however, that the more recent ARPES data, although equally broad in energy, do not seem to show this downward dispersing feature (11).

In the past 10 y, there have been great advances in the study of spin liquids. (For a review, see refs. 13 and 14.) Originally, Anderson (15) and Fazekas and Anderson (16) proposed a spin liquid state for the triangular lattice due to frustration. Now we have strong evidence to support the spin liquid state in at least two cases: the organic compounds close the the Mott transition and in certain Kagome lattices. In light of our recent experience, we can say that existing data are pointing strongly toward a quantum spin liquid state for the 1T-TaS_2 Mott insulator. Due to the rather low value of the sheet resistance, it is generally believed that disorder effects in the undoped samples are small enough that we can rule out an insulator due to Anderson localization. There has been no report of further lattice distortion or phase transition below 200 K, but, in principle, long-range order could set in immediately at 200 K. However, a long-range ordered SDW at wave vector Q will induce a charge density wave order at $2Q$, but these new diffraction spots have not been seen. Furthermore, the ARPES should show clearly the back-folded band instead of a broad smear (10, 11). Finally, since the initial submission of this paper, we have learned that NMR and μSR (spin resonance) data indeed rule out static magnetic moments (17, 18). We therefore believe long-range AF or SDW is unlikely. Instead, 1T-TaS_2 may be an example of the elusive quantum spin liquid.

There is, however, one remaining caveat. Up to now, we have not discussed the issue of interlayer coupling, which can introduce complications for our interpretations. Above 200 K, the ICDW are stacked in an ABC pattern, leading to a tripling of the unit cell in the c direction. However, below 200 K, the stars of David are stacked directly on top of each other to form bilayers. These bilayers are stacked randomly or in an incommensurate fashion (18, 19). The doubling of the unit in the bilayer means that we now have an even number of electrons per unit cell, and a spin liquid ground state may or may not survive, depending on which of the following two options are realized. The first option is for the spins to form singlets between the unit cells. This state no longer fits the definition of a spin liquid, because it is topologically trivial and is smoothly connected to an ordinary band insulator. It is more analogous to ladder compounds, which are still interesting in their own right (20). Indeed, a number of papers (21) have even suggested that the interlayer hopping dominates over the intralayer hopping, and the system forms one-dimensional metals. This point of view is supported the local density approximation with Coulomb repulsion (LDA + U) calculations (21). However, we find this scenario unlikely, in view of the conclusion by Rossnagel and Smith (9) that the split-off bands are mainly xy and $x^2 - y^2$ orbitals, which, unlike the z orbital, have weak interlayer overlap. In contrast, Durancet et al. (21) first ignore spin–orbit coupling and split off an isolated band at the Fermi surface using the LDA+U approximation. The z orbital is strongly admixed and gives a strong interlayer hopping. We believe it is more appropriate to first construct the best single-particle orbital including spin–orbit coupling before turning on U and that the conclusion of Durancet et al. (21) may be an artifact of the LDA+U approximation.

It is important to remark that having a paramagnetic ground state with an odd number of electrons per unit cell is a sufficient but not necessary condition for a spin liquid. The reason is that a second option (other than interlayer singlet formation) is

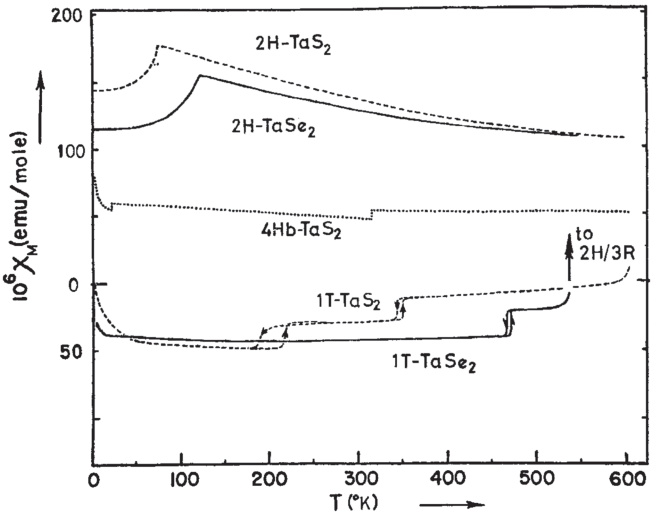

Fig. 2. The molar magnetic susceptibility (χ_M) versus temperature (T) for TaS_2 and T_c with different lattice structures. The background diamagnetic term has not been subtracted. The data are taken from ref. 3.

possible if the spin liquid on each layer is fully gapped. In this case, a sufficiently weak interlayer hopping will not be able to destroy the topological protection of the spin liquid state, as the transition to a topologically trivial state necessarily involves a gap closing. Thus, it is possible for a gapped spin liquid to survive as the true ground state even in the presence of some amount of interlayer coupling. On the other hand, even if the spin liquid is not the true ground state, for small interlayer coupling, there may exist a finite temperature range where the physics is dominated by that of the single-layer spin liquid. There is evidence against the formation of dominant interlayer singlet from the $1/T_1$ data of ref. 17. They do not find an exponential decay with temperature as the dominant interlayer singlet model would imply. Instead, they find a T^2 behavior that may support a Dirac-type spin liquid in the temperature range between 50 K and 200 K. Below 50 K, the system becomes inhomogeneous and hard to interpret.

The purpose of this paper is to bring the exciting possibility of finding spin liquid physics in 1T-TaS_2 to the attention of the community and discuss what kind of spin liquids are consistent with existing data. We also discuss further experiments that can be done to probe this new state of matter. To this end, it will be interesting to look at ultrathin crystals in addition to bulk samples. For example, the $\sqrt{13} \times \sqrt{13}$ structure in free-standing trilayer crystals has been reported to be more robust than in the bulk crystal, being stable even at room temperature (22). It will also be interesting to grow atomically thin samples such as monolayers by molecular beam epitaxy (MBE).

Possible Spin Liquid States in TaS_2

A key feature of spin liquid is the appearance of "fractionalized" excitations, such as particles that carry spin 1/2 but not charge. These particles have been called spinons (15). The spinons can be fermions or bosons. Spin liquids can be divided into two broad classes, gapped or gapless. The gapless spin liquids generically have fermionic spinons, which may form a Fermi surface or Dirac nodes (23). The spinon Fermi surface is characterized by a mass corresponding to a hopping matrix element of order J, the exchange energy. A linear term in the heat capacity with coefficient γ given by this mass has been seen experimentally (24). Theoretically, gauge fluctuation is predicted to convert the linear term to $T^{2/3}$ power, but it has proven difficult to distinguish between the two over the limited temperature range available in the experiment so far. A second key signature is a linear T term in the thermal conductivity, which is usually observed only in a metal (25). It is widely believed that the spin liquid observed in the organics belongs to this type (13, 14). Interestingly, the heat capacity of 1T-TaS_2 shows a linear intercept with small upturn at low temperatures (26). Nevertheless, we believe this observation does not constitute evidence for a spinon Fermi surface for the following reason. The coefficient of this linear term is about 2 mJ/mol·K^2, about 4 times that of copper. This coefficient corresponds to a Fermi energy of about 0.16 eV; we note that the bandwidth of the band at the Fermi level, from band theory, is considerably narrower than this (9). Because we expect J to be at least several times smaller than the bandwidth, the observed γ is much too small to be due to a spinon Fermi surface. It is probably due to the impurity moments seen in the spin susceptibility, forming a random singlet-type state. (In the organics, the γ is several tens in units of mJ/mol·K^2 (24) and corresponds to J of 250 K.) Thus, the heat capacity data effectively rule out a spinon Fermi surface.

A second kind of gapless spin liquid is the Dirac spin liquid. Here the spinons are gapless at certain nodal points and follow a linear spectrum around these points. There are two versions of Dirac spin liquid, U(1) or Z2, depending on whether there exists gapless gauge fields or not. In the latter case, the density of states is linear in energy, giving rise to T^2 heat capacity and NMR relaxation rate $1/T_1$. In the former case, gauge fluctuations may lead to some modifications of these power laws. As noted earlier, the NMR data seem to point to the Dirac spin liquid over a finite temperature range.

Next we come to the gapped spin liquids. In mean field theory, the most common description is one with gapped bosonic spinons (27) together with gapped visons (28); these are called $Z2$ spin liquids, and the visons are the "vortices" of the Z2 gauge fields. We note that, in two dimensions, $U(1)$ gapped spin liquid is not allowed, because the compact $U(1)$ gauge field will lead to confinement. A fermionic mean field theory can also lead to a $Z2$ state with fermionic spinons (29, 30). However, this state is smoothly connected to the one with bosonic spinons, because a bosonic spinon can bind with a vison to form a fermionic spinon, and it is a question of energetics as to whether the low-energy spinons are fermions or bosons. There are more exotic possibilities, but our expectation is that, without further breaking of symmetry, the $Z2$ gapped spin liquid is likely the only common possibility. This state is characterized by low-energy excitations that are gapped spinons (fermions or bosons) and visons. To explain the spin susceptibility data, we will have to argue that the background subtraction in Fig. 2 is such that the susceptibility has dropped to a small value below the first-order transition at 200 K; this means that the spin gap and the exchange scale J are above 200 K. This argument will allow us to get around the issue of the absence of the Curie–Weiss law due to local moments.

There is one more possibility of a gapped spin liquid that has been widely discussed for triangular lattice, and that is the chiral spin liquid (31). This state spontaneously breaks time-reversal symmetry. There should be an easily detected Ising-type transition at a finite temperature, but one could argue that it is not accessible because its temperature scale is above the first-order transition at 200 K. The chiral spin liquid has many observable consequences that have been widely discussed, including Kerr rotation, chiral spin edge modes, and spontaneous quantized thermal Hall conductivity. The absence of signatures of time-reversal symmetry breaking in the μSR data makes this possibility unlikely.

We note that recent numerical work using density matrix renormalization group (DMRG) and variational Monte Carlo methods supports the existence of a region of spin liquid in a $J_1 - J_2$ Heisenberg model on a triangular lattice where the next nearest neighbor (NNN) J_2 is in the range $0.08 < J_2/J_1 < 0.15$ (32–36). Currently, many groups find it difficult to distinguish between the $Z2$ spin liquid, the chiral spin liquid, and the $U(1)$ Dirac spin liquid. These states seem to have very similar energies. It is also important to remember that our system has strong spin–orbit coupling. Consequently, there is no $SU(2)$ symmetry for the pseudospin, and the Heisenberg model is not a good starting model. We expect anisotropic exchange terms. We also expect ring exchange terms, because the system appears to be quite close to the Mott transition. Thus, the phase space may be quite large to support some form of spin liquid.

Effect of Doping or Pressure and Further Experimental Consequences

Next we discuss what is known experimentally when the system is doped or when pressure is applied. It has been reported that 1% Fe doping destroys the CCDW state (37). A Fermi surface appears near the Γ point above this doping level (11), and superconductivity with about 3 K T_c appears. Further doping creates Anderson localization and kills the superconductivity. With a small amount of pressure of 1 GPa, the CCDW is destroyed and replaced by the ICDW, which is, in turn, destroyed with a pressure of 7 GPa (38). The state is metallic and superconducting, with T_c about 5 K everywhere above 4 GPa. We summarize the low-temperature state in the pressure and doping concentration plane in Fig. 3.

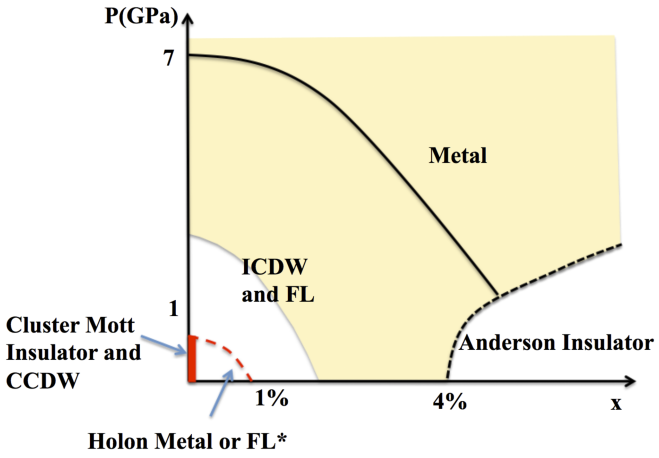

Fig. 3. Schematic phase diagram of the low-temperature state in the pressure *P* and the doping concentration (*x* per Ta) plane. Red dashed line marks a first-order phase transition separating a CCDW and an ICDW. The solid red line marks the undoped spin liquid state formed out of the cluster Mott insulator, the cluster being the star of David shown in Fig. 1. The solid black line separates the ICDW phase and a normal metal phase. The ICDW phase is a Fermi liquid (FL) metal satisfying the Luttinger volume of approximately $1/13 + x$ of the original Brillouin zone volume. Dotted black line marks the onset of Anderson localization due to dopant disorder and is highly schematic. The yellow region denotes superconducting ground state with T_c of $\sim$3 K to 5 K.

It is worth noting that 1% Fe doping is actually a rather large doping in our effective Hubbard model of clusters, because it corresponds to 13% doping per cluster. Thus, it is not surprising that the state looks like a conventional Fermi liquid, with a small and round Fermi surface near the Γ point that presumably obeys the Luttinger volume (11). (Strictly speaking, there is no Luttinger theorem for an incommensurate state, but we can use the nearby commensurate approximate of $\sqrt{13} \times \sqrt{13}$ state to estimate the Fermi surface volume.) The observed superconductivity seems to grow out of this state and extend beyond a pressure where the ICDW is destroyed. Thus, we think this superconductivity may be quite conventional. From the point of view of seeking exotic physics, the most interesting region is the small lower left corner in Fig. 3 where the CCDW order is present. Unfortunately, we are not aware of doping data below 1% carrier per Ta. This region of low dopant density is prone to Anderson localization. Thus, doping by substitution in the plane is likely to create too much disorder. It will be good to attempt doping by intercalation or, even better, by gate in a thin sample. (Focusing on monolayer or trilayer will also help us get around the issue of interlayer coupling.) Indeed, limited data in the very lightly doping range showing metallic-like behavior have been reported by gate doping (39). Here we describe the possible states that result from doping without disorder, and discuss some experimental consequences. Soon after the introduction of the concept on spinons (40) that carry spin and no charge, Kivelson et al. (29) pointed out that an analogous situation may obtain for the doped holes, which carry charge and no spin. These doped holes are called holons. For very small doping, a natural state is a Wigner crystal of holons (41). Here we focus on states that do not break translation symmetry that may emerge with sufficient doping.

Doping a *Z2* Gapped Spin Liquid. In the mean field description with bosonic spinons, the simplest case is that fermionic holons form a Fermi surface with an area corresponding to y, which is defined as the density of dopant per cluster ($y = 13x$ if x is dopant per Ta). For bipartite lattices, there is an additional quantum number corresponding to the A, B sublattice occupied predominantly by the holons (42); for the triangular lattice, this is not the case, and the volume of the Fermi surface corresponds to y spinless fermions. This state has been called the holon metal.

A second possibility is that a holon binds with a spinon to form a physical hole, which, in turn, forms a Fermi surface. Because the hole carries spin, the volume is smaller than the holon metal case by a factor of 2. This state has been called FL* because it behave like a Fermi surface but does not obey the conventional Luttinger theorem with $1 + y$ electrons per cluster (43).

A third possibility is that the state is a superconductor. In mean field theory, this possibility emerges most clearly when the spinons are described as fermions. In this case, the holons are bosons, which will condense and form a conventional superconductor; this is the resonating valence bond (RVB) route to superconductivity as envisioned by Anderson (40).

The holon metal and the FL* are clearly exotic metals, because they are metallic ground state, which dramatically violates Luttinger theorem due to the fact that the Fermi surface volume is y or $y/2$ as opposed to $1 + y$. The physical hole or electron excitation is gapped in the holon metal (because the spinon is gapped) but is gapless in FL*. This distinction will show up clearly in ARPES and tunneling. The FL* will show a Fermi surface, but the holon metal will appear as gapped. Nevertheless, in a clean enough sample, the holon metal will exhibit quantum oscillations. In a multilayer system, the stacked holon metal is insulating in the direction perpendicular to the layer (because only a physical hole can hop between planes), whereas the FL* state is metallic. As discussed earlier, the two states can be distinguished by the size of the Fermi momentum, k_F, which can, in principle, be measured via Koln anomaly or via Friedel oscillations by scanning tunneling microscope (STM) imaging (44).

Doping the Dirac Spin Liquid. The Dirac spin liquid can be $Z2$ or $U(1)$. The former will have gapped visons, whereas the latter has dissipative gapless gauge photons. A natural consequence of doping the Dirac spin liquid is that the bosonic holons condense, resulting in a nodal superconductor. Alternatively, the holons can bind with some of the spinons and form a Fermi surface of volume $y/2$, just like the FL* phase in case 1. The leftover spinons may form their own spinon Fermi surface.

Doping a Chiral Spin Liquid. The natural consequence is a gapped superconductor, which breaks time-reversal symmetry. This has been much discussed, but the effect of spin–orbit coupling has not been explored. The FL* is also a possibility.

Finally, if the ground state of the bulk crystal turns out to be an interlayer singlet, this situation is analogous to that in the ladder compounds in the context of cuprates, and it is possible that doping will induce interlayer pairing of the doped holes, leading to superconductivity (20).

Is there any unique signature of the spin liquid itself? It has been proposed that the undoped $Z2$ gapped spin liquid may show dramatic phenomena when placed in contact with superconductors or magnets (45, 46). For example, Senthil and Fisher (45) pointed out that, if the spin liquid Mott insulator is used as the insulator barrier between two conventional superconductors in an S-I-S structure, this structure may exhibit a charge e Josephson effect. The requirement is that the spatial transition between the superconductor and the spin liquid has to be smooth enough to avoid confinement at the interface. Ordinarily, this is a tall order. However, the 1T-TaS_2 offers a special opportunity in that the superconductors can be created by doping; this can be achieved by gating, which can produce a smooth transition between the superconducting state and the insulating state.

In summary, we believe existing experimental data strongly point to 1T-TaS_2 as an example of a spin liquid state, formed out of a cluster Mott insulator. The lightly doped state is likely an

exotic metal with unusual experimental consequences, some of which we discussed. Ultrathin samples are particularly attractive, because they help avoid the complications of interlayer coupling and can be doped by gating to minimize disorder.

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1. Novoselov KS, et al. (2005) Two-dimensional atomic crystals. *Proc Natl Acad Sci USA* 102:10451–10453.
2. Klemm RA (2011) *Layered Superconductors*, International Series of Monographs on Physics (Oxford Univ Press, New York), Vol 1.
3. Wilson JA, Di Salvo FJ, Mahajan S (1975) Charge-density waves and superlattices in the metallic layered transition metal dichalcogenides. *Adv Phys* 24:117–201.
4. Lu JM, et al. (2015) Evidence for two-dimensional Ising superconductivity in gated MoS2. *Science* 350:1353–1357.
5. Xi X, et al. (2015) Ising pairing in superconducting NbSe2 atomic layers. *Nat Phys* 12:139–143.
6. Saito Y, et al. (2015) Superconductivity protected by spin valley locking in ion-gated MoS_2. *Nat Phys* 12:144–149.
7. Di Salvo FJ, Graebner JE (1977) The low temperature electrical properties of 1T-TaS2. *Solid State Commun* 23:825–828.
8. Fazekas P, Tosatti E (1979) Electrical, structural and magnetic properties of pure and doped 1T-TaS2. *Philos Mag B* 39:229–244.
9. Rossnagel K, Smith NV (2006) Spin-orbit coupling in the band structure of reconstructed 1T TaS_2. *Phys Rev B* 73:73106.
10. Perfetti L, Gloor TA, Mila F, Berger H, Grioni M (2005) Unexpected periodicity in the quasi-two-dimensional Mott insulator 1T- TaS_2 revealed by angle-resolved photoemission. *Phys Rev B* 71:153101.
11. Ang R, et al. (2012) Real-space coexistence of the melted Mott state and superconductivity in Fe-substituted 1T-TaS_2. *Phys Rev Lett* 109:176403.
12. Perfetti L, et al. (2006) Time evolution of the electronic structure of 1T-TaS_2 through the insulator-metal transition. *Phys Rev Lett* 97:067402.
13. Lee PA (2008) An end to the drought of quantum spin liquids. *Science* 321:1306–1307.
14. Balents L (2010) Spin liquids in frustrated magnets. *Nature* 464:199–208.
15. Anderson PW (1973) Resonating valence bonds: A new kind of insulator? *Mater Res Bull* 8:153–160.
16. Fazekas P, Anderson PW (1974) On the ground state properties of the anisotropic triangular antiferromagnet. *Philos Mag* 30:423–440.
17. Klanjsek M, et al. (2017) A new high-temperature quantum spin liquid with polaron spins. arXiv:1704.06450.
18. Kratochvilova M, et al. (2017) The low-temperature highly correlated quantum phase in the charge-density-wave 1T-TaS_2 compound, New Physics Journal, *Quantum Mater*, in press.
19. Ishiguro T, Sato H (1991) Electron microscopy of phase transformations in 1T-TaS_2. *Phys Rev B* 44:2046.
20. Rice TM (1997) *t-J* ladders and cuprate ladder compounds. *Zeit für Physik B* 103: 165–172.
21. Durancet P, Millis AJ, Marianetti CA (2014) Three-dimensional metallic and two-dimensional insulating behavior in octahedral tantalum dichalcogenides. *Phys Rev B* 90:045134.
22. Sakabe D, Liu Z, Suenaga K, Nakatsugawa K, Tanda S (2017) Direct observation of mono-layer, bi-layer, and tri-layer charge density waves in 1T-TaS_2 by transmission electron microscopy without a substrate. *New Phys J Quantum Mater* 2:22.
23. Lee PA, Nagaosa N, Wen XG (2006) Doping a Mott insulator: Physics of high-temperature superconductivity. *Rev Mod Phys* 78:17–85.
24. Yamashita S, et al. (2008) Thermodynamic properties of a spin-1/2 spin-liquid state in a κ-type organic salt. *Nat Phys* 4:459–462.
25. Yamashita M, et al. (2010) Highly mobile gapless excitations in a two-dimensional candidate quantum spin liquid. *Science* 328:1246–1248.
26. Benda JA (1974) Optical, electrical-transport, and heat-capacity studies of the solid solutions $Ti_xTa_{1-x}S_2$, $Zr_xTa_{1-x}S_2$, and $Ti_xNb_{1-x}Se_2$. *Phys Rev B* 10:1409–1420.
27. Read N, Sachdev S (1990) Spin-Peierls, valence-bond solid, and Néel ground states of low-dimensional quantum antiferromagnets. *Phys Rev B* 42:4568–4589.
28. Senthil T, Fisher MPA (2000) Z_2 gauge theory of electron fractionalization in strongly correlated systems. *Phys Rev B* 62:7850–7881.
29. Kivelson SA, Rokhsar DS, Sethna JP (1987) Topology of the resonating valence-bond state: Solitons and high-Tc superconductivity. *Phys Rev B Condens Matter* 35: 8865–8868.
30. Baskaran G, Zou Z, Anderson PW (1987) The resonating valence bond state and high-Tc superconductivity–A mean field theory. *Solid State Commun* 63:973–976.
31. Kalmeyer V, Laughlin RB (1987) Equivalence of the resonating-valence-bond and fractional quantum hall states. *Phys Rev Lett* 59:2095–2098.
32. Mishmash RV, Garrison JR, Bieri S, Xu C (2013) Theory of a competitive spin liquid state for weak Mott insulators on the triangular lattice. *Phys Rev Lett* 111:157203.
33. Zhu Z, White SR (2015) Spin liquid phase of the $S = \frac{1}{2} j_1 - j_2$Heisenberg model. *Phys Rev B* 92:41105.
34. Hu WJ, Gong SS, Zhu W, Sheng DN (2015) Competing spin-liquid states in the spin-$\frac{1}{2}$ Heisenberg model on the triangular lattice. *Phys Rev B* 92:140403.
35. Hu WJ, Gong SS, Sheng DN (2016) Variational Monte Carlo study of chiral spin liquid in quantum antiferromagnet on the triangular lattice. *Phys Rev B* 94:075131.
36. Iqbal Y, Hu W-J, Thomale R, Poilblanc D, Becca F (2016) Spin liquid nature in the Heisenberg J_1-J_2 triangular antiferromagnet. *Phys Rev B* 93:144411.
37. Li LJ, et al. (2012) Fe-doping-induced superconductivity in the charge-density-wave system 1T-TaS_2. *Europhys Lett* 97:67005.
38. Sipos B, et al. (2008) From Mott state to superconductivity in 1T-TaS_2. *Nat Mater* 7:960–965.
39. Yu Y, et al. (2015) Gate-tunable phase transitions in thin flakes of 1T-TaS_2. *Nat Nanotechnol* 10:270–276.
40. Anderson PW (1987) The resonating valence bond state in La_2CuO_4 and superconductivity. *Science* 235:1196–1198.
41. Jiang HC, Devereaux T, Kivelson SA (2017) Holon Wigner crystal in a lightly doped Kagome quantum spin liquid. arXiv:1702.06577.
42. Shraiman BI, Siggia ED (1988) Mobile vacancies in a quantum Heisenberg antiferromagnet. *Phys Rev Lett* 61:467–470.
43. Senthil T, Sachdev S, Vojta M (2003) Fractionalized Fermi liquids. *Phys Rev Lett* 90:216403.
44. Mross DF, Senthil T (2011) Charge Friedel oscillations in a Mott insulator. *Phys Rev B* 84:041102(R).
45. Senthil T, Fisher MPA (2001) Detecting fractions of electrons in the high T_c cuprates. *Phys Rev B* 64:214511.
46. Barkeshli M, Berg E, Kivelson S (2014) Coherent transmutation of electrons into fractionalized anyons. *Science* 346:722–725.